Microbeam Analysis of Yellow Sand Dust Particles

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Introduction

Yellow sand dust particles from the Asian continent sometimes cause turbid conditions in Japan, especially in spring, which is known as a Kosa event. The particles are carried from the Yellow River basin and deserts, which is known as a natural phenomenon. The particles are principally composed of minerals and their diameters are around 4 μm. Recently, it is regarded as an environmental problem due to forest reduction, soil degradation and desertification rather than a natural seasonal phenomenon. Furthermore, these particles are sometimes deformed by mixing with anthropogenic aerosols during transport and affects agricultural production, climate, living condition and human health. Therefore, physical and chemical changes during transport as well as mechanism of formation, growth and transport have to be known. For better understanding of these mechanisms, analysis of single particles is indispensable. For this purpose, we developed a microbeam analysis system with a spatial resolution better than 1 μm. The combination of PIXE, RBS and off-axis STIM methods enabled simultaneous analysis for hydrogen to metal elements and revealed the chemical composition of these particles. In this study, we applied the system to analyze yellow sand dust particles and investigated their properties.

Sampling

Aerosol particles were collected at the campus of Nagasaki University (32.78°N, 129.87°E; 20 m) on 9,10 and 19 May 2005. Nagasaki University located at Kyusyu Island of western part of Japan, where yellow sand dust particles largely affects the daily life.

Recently, fine particles whose sizes are in the range of 0.3-1.0 μm in diameter occasionally cause significant turbid condition over the Nagasaki area. Since yellow sand dust particles are mostly larger than 1.0 μm, these fine particles are mainly anthropogenic aerosols and not the components of yellow sand dust particles[1,2]. To collect true yellow sand dust particles, sampling was carried out when concentration of coarse particles increases and that of fine particles shows usual atmospheric condition by using an optical particle counter (OPC). On 9 and 10 May, coarse particles showed higher concentration, indicated Kosa events and fine particle showed as usual condition. On 19 May, concentrations of fine and coarse particles were low. Sampling times were 6 hr. In total 3 samples were collected. Aerosol particles were impacted on a thin polycarbonate film[3] at flow rates of 1 *l*/min (face velocity: 530cm/sec). The effective 50% cut-off diameter is estimated to be ~2 μm and sufficient for collection of yellow sand dust particles. Elemental concentrations and ratios in the polycarbonate film were obtained by fitting the RBS spectrum with the SIMNRA software[4]. Thickness of the film was estimated to be less than 1 μm which is thicker than the previous study. The thin polycarbonate film allows analyzing light elements of the particles.

Analysis

Analysis was carried out using the microbeam analysis system at Tohoku University. Technical details of the microbeam and analysis system were presented in previous ppers[5-7] and further development was carried out for the efficient analysis of single aerosol particles. For multimodal analysis, two X-ray detectors for PIXE analysis and three charged particle detectors for RBS and STIM are mounted simultaneously[8].

Results

Simultaneous PIXE/RBS/off-axis STIM analysis employed for scanning areas of 25×25 μm^2 and 100×100 μm^2. Yellow sand dust particles were uniformly distributed, thus direct STIM measurements for defining the analysis area were not needed. Energy of the proton beam was 3 MeV and beam spot size was 1×1 μm^2 with a beam current of 50~100 pA. Total accumulated charge was around 0.2 μC.

Quantitative PIXE analysis was performed using the GeoPIXEII software[9]. After generating the elemental maps, individual particles were selected from these maps and PIXE, RBS and off-axis STIM spectra were extracted. Elemental concentrations of these particles for elements heavier than Na were deduced from fitting of the extracted PIXE

spectra. Carbon and oxygen were quantified by analyzing the extracted RBS spectra. Concentration of carbon and oxygen were derived from peak yields which were calibrated by measuring peak yields from Mylar films of known thickness. Quantitative analysis of hydrogen was carried out by analyzing the extracted off-axis STIM spectra. For the quantitative analysis of hydrogen, intensities of hydrogen were calibrated by measuring hydrogen yields from Mylar films of different thickness. The relation between hydrogen peak yield and hydrogen quantity was linear and was used to calibrate the quantitative analysis of hydrogen. After quantification of these particles, the chemical composition of each particle was obtained.

More than 100 particles were analyzed and Na, Mg, Al, Si, S, Cl, K, Ca, Ti, V, Cr, Mn, Fe, Ni, Cu, Zn and Br elements were quantified. Figure 1 shows typical elemental maps of yellow sand dust particles collected on 11 May, 2005, when high concentration of coarse particles was observed. The scanning area was 100×100 μm^2 and total accumulated charge was 0.21 μC. Distribution of aluminum equals to the one of silicon. Chlorine, potassium, calcium, carbon and hydrogen also show similar distribution. Oxygen distributes on the same part of these particles. Aluminum, silicon, calcium, iron and light elements are major components. Numbers of oxygen atoms of these particles is around eight times higher than that of mineral elements. It is apparent that these particles manly existed as oxide and were mainly originated form soil dust. These particles also contained carbon and hydrogen and might be absorbed organic carbon during transportation. In the contrast, the particles collected on 19 May when atmosphere showed usual condition, two kinds of major elemental groups, which correspond to marine and soil aerosols, were observed. Half of the particles are estimated to be marine aerosols.

Figure 2 shows the ternary diagram for calcium, silicon and aluminum. In this diagram, marine particles, which sodium and calcium are main components, were excluded. The distribution of the particles which were collected on 10 and 11 May were very similar, except for calcium-rich components. On the other hand, the particles collected on 19 May show lower calcium concentration. These facts correspond to the difference of the soil components of Asian continent and Japan Island. Correlation function between calcium and chlorine is shown in Fig. 3 for these particles. As calcium increases, chlorine also increases for the particles collected on 11 May. The particles are supposed to contain $CaCl_2$ particles. Backward trajectory analysis using NOAA HYSPLIT[10] indicated that the particles which came on 11 May took almost the same route from Chinese coast to

Nagasaki with a low velocity as the ones came on 10 May. Amount of sodium on 11 May is also larger than that of 10 May. It implies that the particles came on 11 May deformed by mixing with marine aerosols more than those came on 10 May. These particles collected on 10 and 11 May contained more sulfur and heavier elements such as manganese and zinc than those collected on 19 May. These particles were transported from Asian continent over the industrial area to Japan and might absorb these elements by mixing with anthropogenic aerosols.

Conclusions

The microbeam system was applied to analysis of yellow sand dust particles. Simultaneous PIXE, RBS and STIM analysis enabled to measure the elements from hydrogen to metal elements and revealed the chemical composition of these particles. The major elements of yellow sand dust particles are silicon and calcium and exist as oxide. The ternary diagram of aluminum, silicon and calcium shows the difference of the origin of the soil components of Asian continent and Japan Island. Correlation of calcium and chlorine is different for each Kosa events, which supposed the difference of transport to Japan Island. Yellow sand dust particles contain more sulfur and heavier elements such as manganese and zinc. These particles were transported from Asian continent over the industrial area to Japan and might absorb these elements by mixing with anthropogenic aerosols. These results could be obtained by analyzing single aerosol particles and was not obtained by the bulk analysis which averages over many particles. Single particle analysis of yellow sand dust will lead to a better understanding of their deformation process during transport.

Acknowledgments

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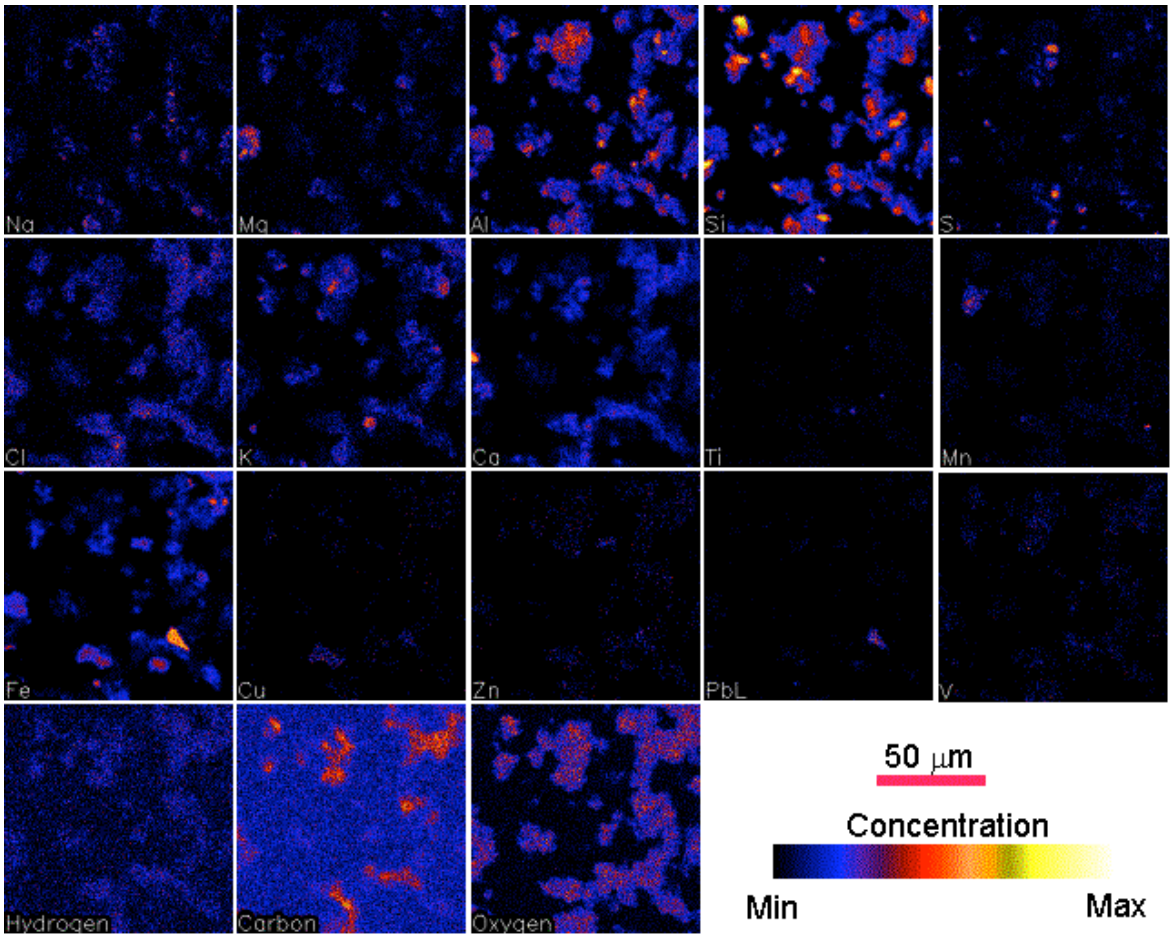

Figure 1. Elemental Distribution images of Yellow Sand Dust Particles collected on 11 May, 2005.

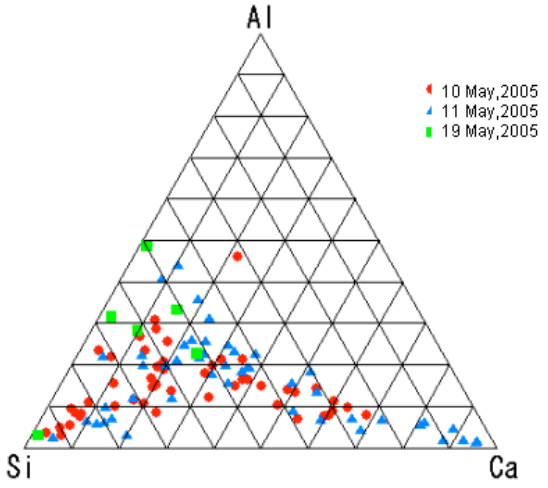

Figure 2. Ternary Diagram of Collected Aerosols.

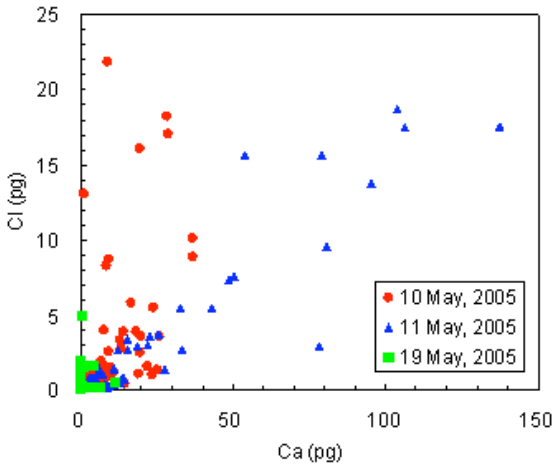

Figure 3. Correlation between Ca and Cl.